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Microplastics in agricultural drainage water: A link between terrestrial and aquatic microplastic pollution

Moritz Bigalke^{a,*}, Milo Fieber^a, Alexandra Foetisch^a, Julien Reynes^b, Peter Tollan^b

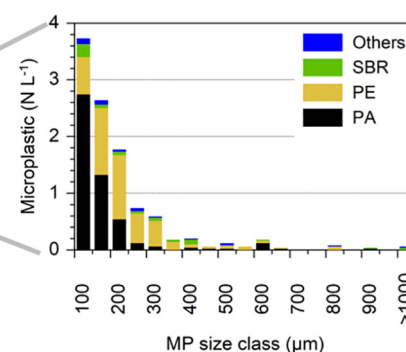
^a Institute of Geography, University of Bern, Hallerstrasse 12, 3012 Bern, Switzerland

^b Institute of Geology, University of Bern, Baltzerstrasse 3, 3012 Bern, Switzerland

HIGHLIGHTS

- Agricultural drainage water from Switzerland contained microplastics.
- The concentrations imply significant transport of microplastics in the soil.
- The findings indicate a transfer between soil and aquatic microplastics.
- The results show relevant inputs of microplastics to local surface waters.
- The fluxes might be relevant for global microplastic modeling.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastic (MP) contamination has been reported to be higher in terrestrial compared to aquatic environments. This is probably due to the fact that plastic items are mostly produced and used in terrestrial environments and have a longer residence time. However, there are several links between the terrestrial and aquatic environments. We analyzed drainage water samples from agricultural soils in the Seeland, a heavily drained agriculturally intensive area in Switzerland for its MP (>100 μm) concentration and composition. We found MP in relevant numbers (mean $10.5 \pm 9.5 \text{ N L}^{-1}$). The polymers were mainly PA and PE, and the size distribution showed an exponential increase with decreasing particle size. The results show considerable MP concentrations in drainage water and imply a transport of MP in soils down to the drainage pipes. Given the large areas drained both in Switzerland and globally, it is proposed that MP leaching from soil can be a significant source of MP to aquatic ecosystems. Such a contribution should be considered when dealing with MP cycling on a local to global scale.

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1. Introduction

Since 1950, about 8300 million metric tons of plastic have been produced worldwide, 79% of which are accumulated either in landfills or in the environment (Greyer et al., 2017). Soils are the main reservoirs of waste plastics, having 4–23× higher plastic concentrations compared to aquatic systems (Horton et al., 2017). However, as the environmental

* Corresponding author.

E-mail address: moritz.bigalke@giub.unibe.ch (M. Bigalke).

compartments are linked, microplastics (MP) can readily exchange between terrestrial to aquatic systems and vice versa (Horton et al., 2017). On the one hand, plastic particles can be transferred from aquatic to soil systems e.g. by flooding or irrigation (Scheurer and Bigalke, 2018; Blasing and Amelung, 2018). On the other hand, terrestrial MP can be transferred to aquatic systems by erosion and runoff (Rezaei et al., 2019; Tagg and Labrenz, 2018; Horton et al., 2017) or with drainage water from drained soils. While water erosion might be more significant in areas with elevated topography, drainage water is more important in flat areas and former wetlands (e.g. Germany, USA; Schultz et al., 2005). Initial studies only considered erosion and runoff when modeling soil to water fluxes of MP, neglecting the contributions from drainage water (Tagg and Labrenz, 2018; Horton et al., 2017; Nizzetto et al., 2016; Hurley and Nizzetto, 2018).

Drained areas are generally under intense human use (otherwise they would not be drained) and host settlements, infrastructure or agriculture. In all these land use systems, plastics are likely to be broadly dispersed, e.g., due to littering and plastic use in agriculture (Blasing and Amelung, 2018; Kawecki and Nowack, 2019). In settlements, soils are mostly sealed and drainage water is treated in sewage systems before release back into the environment. However, in agriculture, drainage water is mostly released to aquatic systems without any treatment. While subsurface drainage is the most common drainage system in temperate zones, surface drainage is more common in the arid and semi-humid areas of the world. In total, open drains and surface drainage make up about 55% of the drained area, while subsurface drainage and vertical drainage cover 38% and 7% respectively (Schultz et al., 2007). Due to the transport of MP in soils (Maass et al., 2017; Rillig et al., 2017; Zubris and Richards, 2005; Wanner, 2021) the plastics dispersed at the soil surface can reach deeper soil sections and eventually the shallow groundwater table. These transport pathways enable MP to be transported to the subsurface drainage system and be directly drained to adjacent surface waters (Wanner, 2021).

Currently, however, there is no information about MP leaching to, and MP concentrations in, drainage water, making it impossible to assess its impact on MP concentration in surface waters. To address this significant gap in knowledge, we analyzed drainage water samples from 11 different subsurface drainage sites at the Swiss Seeland to answer the following questions:

- 1) Are the MP present in agricultural soils transported sufficiently in order to reach the drainage water system?
- 2) Does drainage water contribute to increased MP concentrations in surface waters?

2. Material and methods

Eleven study sites were selected in the Swiss Seeland (Fig. 1). The Seeland is an intense agricultural area with a high proportion of intense vegetable production and a high use of mulch foil, mulch tunnels and other plastic items. From the eleven sites, eight were equipped with a drainage systems where the water is either constantly passively released through an open system or first collected in a collection basin and then passively released (Fig. 1). The other three sites were equipped with a newer type of drainage system where the water is collected in a collection basin and then automatically pumped to surface waters, when the water level in the basin reach a certain level. In the latter system, surface water which has not been transported through the soils can also contribute to the water in the basin. Five of the sites were sampled at two different times (31.01.2020 and 03.05.2020) to test the temporal variability. Both sampling dates were set at times with considerable precipitation. The sum of precipitation of the 7 and 3 days before sampling were 31.6 mm and 16 mm at January 31, and 43.1 mm and 12.7 mm at May 03, respectively. In total we analyzed 19 samples from 11 sites.

At the study sites with flowing water (F1-F8), the water was sampled directly into a 3 L glass container. When the samples were taken from a collection basin (S1-S3), they were sampled with a pre-cleaned metal bucket and then transferred to a 3 L glass container (Fig. 1). When taking a sample with a bucket, the light MP (e.g. PE, PA) floating on the surface of the collection basin might be favored compared to heavy MP (e.g. PET, SBR) which will sink to the bottom of the basin.

The glass container was pre-cleaned three times with Milli-Q® water and three times with the drainage water directly at the site. The samples were further processed following the protocol of Cabernard et al. (2016). The samples were filtered through a membrane filter with a pore diameter of 8 µm (Whatman®, Cellulose Nitrate). The filtered material was recovered by placing the filter upside down, backwashing with 15 mL of 60% H₂SO₄ (density 1.5 g cm⁻³) and collecting the rinsing solution. After exposure to the acid (30 min) to oxidize organic material, the samples were centrifuged (3000 RPM, 30 min, 20 °C) to separate the MP from the inorganic material by density separation. The supernatant containing the MP was filtered through an Anodisc filter (Whatman®, 0.2 µm), which was subsequently washed with MilliQ water and dried before Fourier transform infrared spectroscopy (FTIR) analysis for chemical characterization and identification. The Anodisc filter is suitable for FTIR analysis in the mid-IR for wavenumbers >1200 cm⁻¹ (Löder et al., 2015).

Each filter was measured with a Bruker Tensor II FTIR coupled to a Hyperion 3000 microscope (Bruker Cooperation, Billerica, MA). Single

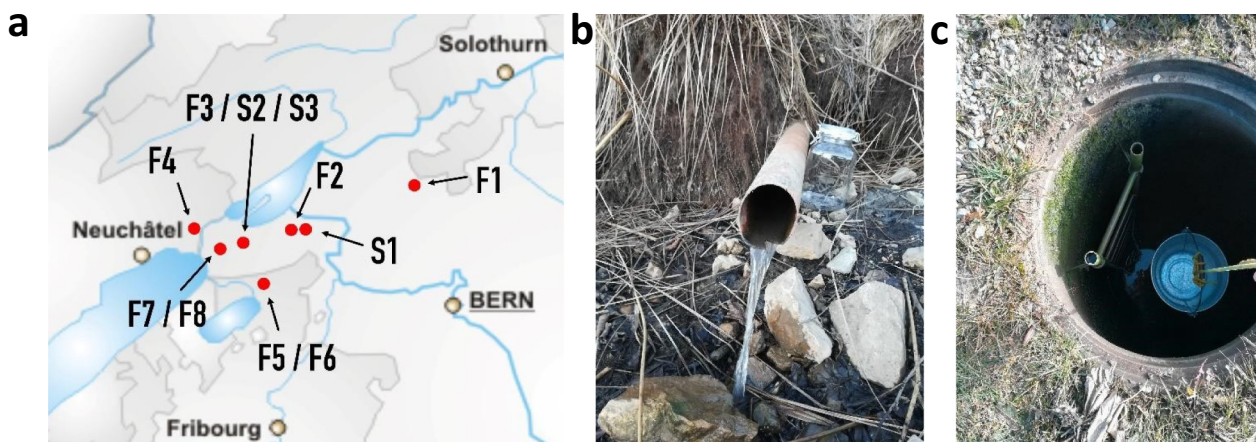


Fig. 1. Sampling sites, a) Location of the sampling sites in Switzerland. Fx corresponds to sampling site where the water flow is passive and constant. Sx corresponds to sampling sites where the drainage water is collected into a basin before being released into surface waters. b) Example for constant drainage at F7 and c) drainage water collection at S3 (while sampling with the metal bucket).

measurements were acquired with a square aperture ($\sim 100 \times 100 \mu\text{m}$) using a $15\times$ cassegrain objective and a liquid nitrogen-cooled mercury cadmium telluride (MCT) detector in transmission mode over the wavenumber range $600\text{--}4000 \text{ cm}^{-1}$. Background measurement were done on clean Anodisc filter with 32 scans and sample measurements with 32 scans and using a resolution of 4 cm^{-1} . The spectra were cut in the range $1300\text{--}3300 \text{ cm}^{-1}$ and baseline corrected (10 iterations, concave rubberband algorithm from OPUS® software).

Chemical identification was achieved by comparing four distinct wavenumber regions ($2980\text{--}2780$, $1800\text{--}1740$, $1760\text{--}1670$, $1480\text{--}1400 \text{ cm}^{-1}$) with the database and protocol from Löder et al. (2015), using the spectrum search tool from OPUS®. Particles were classified as MP if hit qualities were >700 . Only particles $>100 \mu\text{m}$ in diameter were analyzed. We tested the effect of the method on polyethylene (PE), polypropylene (PP) and polyvinylchloride (PVC) MP (about 1 mm in diameter) and found no visual change (under the microscope) of the particles following the acid treatment, consistent with the results of Cabernard et al. (2016). To prevent contamination of the samples, most preparation steps were done under a laminar flow clean hood with HEPA (H14) filtered air and all lab equipment was rinsed with Milli-Q® water and ethanol before use. To test for possible contamination, blanks (2.5 L Milli-Q® water) were processed together with the samples and gave a mean count of $0.8 \text{ particles L}^{-1}$. The detection limit of our method ($\text{mean}_{\text{blank}} \pm 3\text{SD}$) was calculated to be $2.2 \text{ particle L}^{-1}$. To determine the particle recovery rate of the method, 10 MP particles (diameter $1\text{--}2 \text{ mm}$) each of PE, PP and PVC were dispersed in water and prepared like the normal samples. Following this, all of the

particles were successfully recovered. One sample was sampled and analyzed in triplicate and one in duplicate. The mean RSD was 54%, probably due to the high heterogeneity of the water samples.

3. Results

The number (N) of MP L^{-1} in the drainage water shows considerable variation, with MP numbers being below detection limit in some samples while in other samples reaching up to 34.6 N L^{-1} (Fig. 2). The mean of the MP number is 10.5 N L^{-1} . The mean relative standard deviation (RSD) of samples sampled two times at the same sampling site but at different times was 72% (range 38–106%), indicating high variability. Polyamide (PA) and PE are the most common polymers in the drainage water (Fig. 2, b). The size distribution of the MP showed a strong increase with increasing particle size, with PA being the most abundant polymer in the smallest size range ($100\text{--}150 \mu\text{m}$) and PE being the most abundant in the size range $150\text{--}300 \mu\text{m}$ (Fig. 2, c).

4. Discussion

4.1. The leaching of MP in soils

The occurrence of MP in the drainage water sheds light on the transport of MP in soils. Depth transport has been reported along preferential flow path (Zubris and Richards, 2005) but was also reported in unsaturated column experiments, without preferential flow (Mitropoulou et al., 2013; Hoggan et al., 2016). Column experiments with spherical

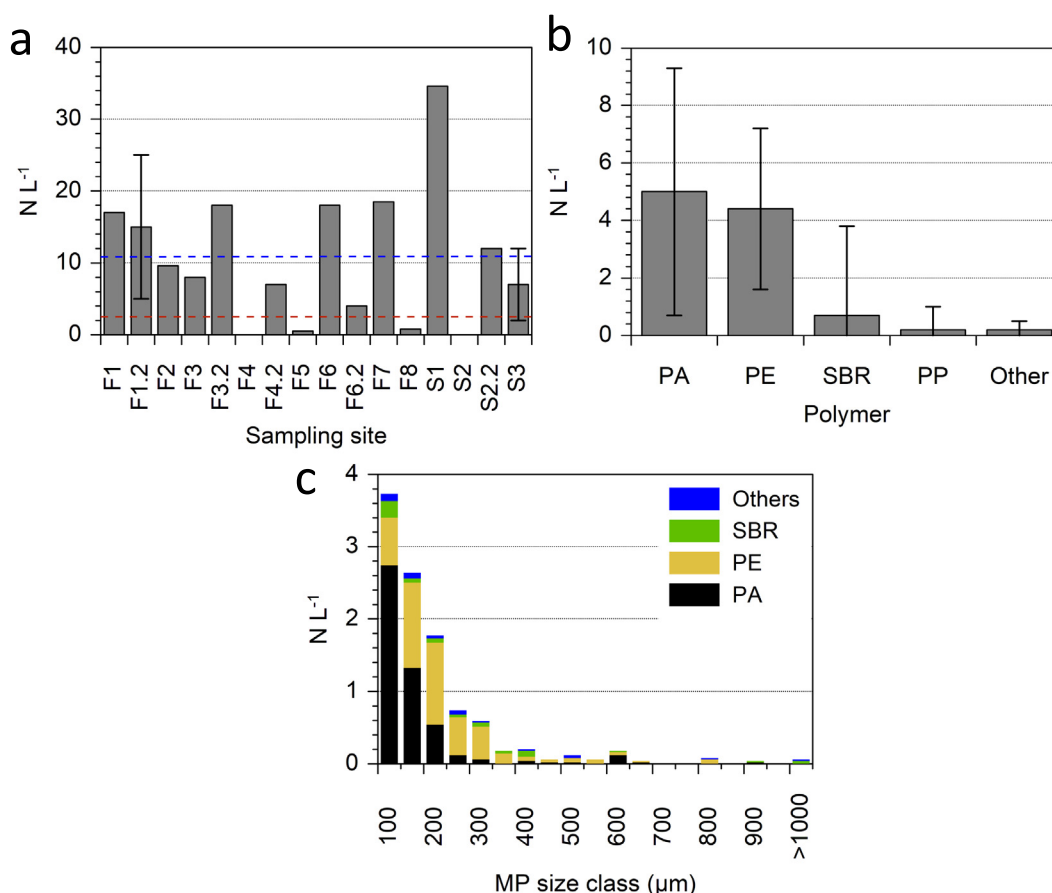


Fig. 2. a) MP numbers in the individual samples. Samples labeled with 0.2 in the name were taken at the second sampling date. Error bars show the standard deviation of replicate samples taken at the same time. The red line show the method limit of detection, the blue line show the mean value b) Mean particle numbers of the individual polymers in the drainage water samples. Error bars refer to standard deviation between all sites. c) The size dependent MP numbers and polymer composition. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

shaped microplastic particles illustrated that the transport is size dependent, that aggregation and deposition of the particles is the major removal mechanism from soil, that these processes depend on the ionic strength of the solution and that a lower pore water saturation resulted in increased retention (Hoggan et al., 2016; Mitropoulou et al., 2013; Wanner, 2021). More advanced studies with differently shaped particles and columns with natural soils or complex fillings illustrated the co-transport of MP with mobile organic particles and the effect of particle shapes (spheres being more mobile than fibers). Furthermore, they demonstrated that biotic factors like earthworm activity can also have a considerable effect on the transport of MP (Lwanga et al., 2017; Maass et al., 2017; Rillig et al., 2017).

Based on those findings plastic transport to groundwater has been assumed (Wanner, 2021). Our data show that this transport occurs under normal environmental conditions under a central European climate and moderate precipitation and that MP can reach drainage pipes and therefore also the shallow groundwater. Furthermore, the size distribution of the MP in the water samples shows that smaller particles ($<350\mu\text{m}$) are much more mobile and reach drainage water while bigger particles are rarely present. From the literature about plastic leaching (Wanner, 2021) it can be assumed that particles $<100\mu\text{m}$, which were not analyzed in this study, will be even more mobile in the soils and will occur in much higher concentrations. Our samples show a considerable variability at sampling at the same time point, which we assume to be due to the small water volume analyzed and the high heterogeneity of MP leaching. The variability between the different sampling dates is even more pronounced and probably due to the difference in precipitation events and water flow in the soils at the different times.

The PE found in the drainage water is consistent with the heavy use of PE for mulch-, greenhouse- and tunnel foils on soils but also the fact that modern drainage pipes are often made of PE (Scarascia-Mugnozza et al., 2011). The PA has no common use in agriculture but is mostly used for textile and packaging production. The most probable sources for this polymer are past sewage sludge applications (Mahon et al., 2017) and littering. Styrene butadiyl rubber (SBR) is a common polymer in tires and probably indicates the presence of tire wear in the water samples, while PP is used for similar applications as PE, just in a lower quantity. As plastic use in agriculture still persists and plastic has a very long residence time in soils, we assume that plastic leaching to drainage waters will continue. Furthermore, the plastic disintegration to MP as well as its transport in soils takes time (Zubris and Richards, 2005; Krueger et al., 2015) so that the MP we find in the drainage water today were probably released to the soil years ago. This means that even if plastic emission to soils is strongly reduced and agriculture change to biodegradable mulch films, a decrease of plastic leaching from soil will only occur after a prolonged transition period.

4.2. Implications of MP leaching from soils to surface waters

The MP concentrations reported in the literature are strongly dependent on the MP sizes analyzed. In some studies Manta nets with a mesh size of $300\mu\text{m}$ are used to sample the MP. This kind of sampling normally generates low MP counts, because only bigger MP are sampled. The number of MP reported increase strongly when other methods are used that consider smaller MP. The different sampling and analytical techniques used make the data of different studies hardly comparable (Triebkorn et al., 2019). The MP concentrations we find in the drainage water samples ($10.5 \pm 9.5\text{ N L}^{-1}$) are considerable, and tend to be higher than the concentrations in Swiss surface waters ($7 \pm 5\text{ N L}^{-1}$, Cabernard et al., 2016; 0.007 N L^{-1} , Faure and Alencastro, 2014) and comparable or higher than most surface water systems (Triebkorn et al., 2019). With these numbers, our data indicate that MP in drainage water might have a considerable influence on surface water concentrations. However, more studies with a wider geographical spread about the MP concentrations in surface and drainage water are needed to

get a better and more general idea on the impact of MP release through drainage water channels.

Depending on the MP concentrations drainage water might contribute considerable to MP release. Taking Switzerland as an example, about 18% ($1.9 \cdot 10^5\text{ ha}$) of the agricultural area are drained, mainly with sub-surface drainage. At these sites, 58–86% of the precipitation goes into the drainage systems, which contributes about $8.9 \cdot 10^8\text{ m}^3$ or 2% to the total runoff in Switzerland (Gramlich et al., 2018; Kobierska et al., 2020; Hürdler et al., 2015). With a rough estimation based on our MP concentrations, this would mean about $9.3 \cdot 10^{12}$ MP particles released from agricultural drainage in Switzerland each year. Worldwide about $1.9 \cdot 10^8\text{ ha}$ of agricultural land are drained, which represents about 13% of the total arable land (Schultz et al., 2005). If we roughly assess the water from drainage systems with a mean global precipitation of about 1000 mm m^{-2} (Adler et al., 2017) and assume a drainage of only about 50% we can assess that about $9.5 \cdot 10^{11}\text{ m}^3$ water are going through drainage systems every year. This is of course only a very rough estimate but illustrates that MP in this water constitutes a considerable flux.

Most of the drainage systems are surface drainage (Schultz et al., 2007) and cannot be directly compared to our data. However, we assume that surface drainage might also have an effect on plastic transport as plastic is not filtered in the soils, but rather directly washed away to surface waters. Most of the drained area is in developed countries with intensive agriculture involving plastic use (Schultz et al., 2007; Schultz et al., 2005). Thus, the MP load might be significant not only for the directly affected rivers and lakes, but also for the global aquatic plastic budget.

5. Conclusions

Our results show a considerable presence of MP in drainage water and imply that MP are leached through the soils to drainage water systems. Given the drained area worldwide, MP from drainage water might contribute to increased MP concentrations in surface waters. Thus, drainage water should be considered when assessing MP fluxes to surface waters. Future research is needed to confirm our findings with a higher number of samples from different locations worldwide.

CRedit authorship contribution statement

Moritz Bigalke: Conceptualization, Funding acquisition, Project administration, Supervision, Writing – original draft. **Milo Fieber:** Conceptualization, Methodology, Investigation, Formal analysis, Data curation, Writing – review & editing. **Alexandra Foetisch:** Methodology, Resources, Writing – review & editing. **Julien Reynes:** Methodology, Investigation, Resources, Writing – review & editing. **Peter Tollan:** Methodology, Investigation, Resources, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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